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**Exotic Charge-Ordered State in  $\text{Bi}_{1-x}\text{Ca}_x\text{MnO}_3$  ( $x > 0.5$ ):  
Spectroscopic Ellipsometry and Raman Scattering Studies\***

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We present the effects of charge-ordering (CO) on the charge-, spin-, and lattice-dynamics of the  $\text{Bi}_{1-x}\text{Ca}_x\text{MnO}_3$  ( $x > 0.5$ ) using spectroscopic ellipsometry (SE)<sup>†</sup> and Raman scattering (RS)<sup>‡</sup> studies. SE has allowed us to study how CO influences the charge- and orbital-degrees of freedom on various length scales by probing the dielectric anisotropy at different temperature and energies. The temperature evolution of the dielectric anisotropy also exhibits interesting similarities to the development of the anomalous lattice dynamics, probed by RS. Additionally, we observe in the antiferromagnetic (AFM) charge-ordered phase the development of a quasielastic Raman response, which has the symmetry of the spin-chirality operator ( $\vec{S}_1 \cdot \vec{S}_2 \times \vec{S}_3$ ). This quasielastic response suggests the presence of strong magnetic fluctuations at finite temperatures in the AFM CO phase. Among the possible sources of this anomalous response are (i) fluctuations of the Mn core spins associated with a canted AFM or spin-chiral phase, and (ii) fluctuations associated with closed-loop charge currents.

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